

REMARKS

Claims 9-16 are pending in this application. By this Amendment, claims 9-12 are amended, claims 1-8 are canceled and new claims 13-16 are added. Claims 9 and 12 are amended to address the teachings of the cited references.

No new matter is added to the application by this Amendment. Support for the amendments to claims 9 and 12 can be found in paragraph [0078] of the specification, as filed. Support for new claims 13-16 can be found in canceled claims 4-8.

I. Rejections Under 35 U.S.C. §103(a)

A. Suhara et al. in view of JP '410

Claims 1, 2, 4, 6, 8, 10 and 11 were rejected under 35 U.S.C. §103(a) as allegedly being obvious over U.S. Patent No. 5,953,204 to Suhara et al. in view of JP 2001-146410 (JP '410). This rejection is respectfully traversed.

In light of the cancellation of claims 1, 2, 4, 6 and 8, the rejection with respect to these claims is considered moot.

Claims 10 and 11 are amended to depend from claim 9. Since claim 9 was not rejected as allegedly being unpatentable over Suhara et al. in view of JP '410, the rejection of these claims has been overcome. Reconsideration and withdrawal of this rejection are respectfully requested.

B. Suhara et al. in view of Sonobe et al.

Claims 1 and 7 were rejected under 35 U.S.C. §103(a) as allegedly being obvious over Suhara et al. in view of U.S. Patent No. 6,258,337 to Sonobe et al. This rejection is respectfully traversed.

In light of the cancellation of claims 1 and 7, the rejection with respect to these claims is considered moot. The subject matter of original claim 7 has been retained in new claim 14,

which depends from claim 9. Since claim 9 was not rejected relying upon Suhara et al., Applicants submit that claim 14 is allowable in view of these references.

Reconsideration and withdrawal of this rejection are respectfully requested.

C. JP '821 in view of JP '410

Claims 1, 2, 4, 6, 8 and 9-11 were rejected under 35 U.S.C. §103(a) as allegedly being obvious over JP 60-211821 (JP '821) in view of JP '410. This rejection is respectfully traversed.

In light of the cancellation of claims 1, 2, 4, 6 and 8, the rejection with respect to these claims is considered moot.

Neither JP '821 nor JP '410, taken singly or in combination, teaches or suggests an electrochemical capacitor comprising a cathode that contains a fibrous carbon material having an electronic conductivity as a constituent material, wherein the fibrous carbon material has a specific surface area of 2000 to 3000 m²/g as recited in amended claim 9.

JP '821 discloses two electric double layer capacitors (i) and (ii), each having a fibrous carbon material as a positive electrode and a granular activated carbon material as a negative electrode. In JP '821, the electric double layer capacitor (i) includes a fibrous carbon material having a specific surface area of 900-1000 m²/g as a positive electrode, and a granular carbon material having a specific surface area of 1400 m²/g as a negative electrode. On the other hand, electric double layer capacitor (ii) of JP '821 includes a fibrous carbon material having a specific surface area of 1500-1600 m²/g as a positive electrode, and a granular carbon material having a specific surface area of 1400m²/g as a negative electrode (see the translation of the description in lines 11-19 and the translation of Table 4 of JP '821 filed with the August 22, 2006 Information Disclosure Statement).

However, it is described in JP '821 that "a specific surface area of positive electrode-side of the polarizable electrode body is smaller than the specific surface area of a negative

electrode side of the polarizable electrode body." (see lines 16-19, page 2 and lines 2-6, page 7 of the attached translation of JP '821). In JP '821, it is also described that "an object of the present invention is to provide an electric double layer capacitor with improved efficiency for forming an electric double layer per unit volume." (see lines 23-25, page 6 of the attached translation of JP '821).

The requirement that a specific surface area of the positive electrode-side polarizable electrode is smaller than that of the negative electrode-side polarizable electrode in the electric double layer capacitor (i) of JP '821 is not satisfied if the specific surface area of the fibrous carbon material is changed to 2000-3000 m²/g as recited in claim 9. Additionally, the requirement that a specific area of the positive electrode-side polarizable electrode is smaller than that of the negative electrode-side polarizable electrode in the electric double layer capacitor (ii) as required in JP '821 is not satisfied if the specific surface area of the fibrous carbon material is changed to 2000-3000 m²/g as required in claim 9. JP '821 thus teaches away from the electrochemical capacitor of claim 9

Further, JP '821 describes:

"For the above reasons, a conventional electric double layer capacitor which uses activated carbon fiber as the polarizable electrode and an organic electrolytic solution as the electrolytic solution has an extremely large specific surface area of 2000 m²/g for both the positive and negative electrodes, and uses activated carbon fibers where a majority of the pore diameters are between 2 and 4 nm. However, activated carbon fibers which have these large characteristics have a disadvantage in that the activation efficiency is extremely low, at approximately 20%." (see lines 11-20, page 6 of the attached translation of JP '821).

Such further teaches one of ordinary skill in the art away from the electrochemical capacitor of claim 9, JP '821 specifically discouraging the use of higher surface area carbon fibers.

Thus, JP '821 teaches one of ordinary skill in the art away from an electrochemical capacitor comprising a cathode that contains a fibrous carbon material having an electronic conductivity as a constituent material, wherein the fibrous carbon material has a specific surface area of 2000 to 3000 m²/g, as recited in claim 9.

The Patent Office acknowledged that JP '821 does not disclose an activated carbon powder that is substantially spherical. The Patent Office introduces JP '410 to allegedly teach an improved activated carbon powder having a spherical shape used in electrochemical capacitors.

Even if JP '410 were to have been combined with JP '821 as alleged by the Patent Office, the subject matter of the present claims would not have been achieved. JP '410 does not remedy the deficiencies of JP '821 detailed above. For example, like JP '821, JP '410 also does not disclose the cathode having fibrous carbon material that has a specific surface area of 2000 to 3000 m²/g.

For at least the foregoing reasons, Applicants respectfully submit that JP '821 and JP '410, taken singly or in combination, fail to teach or suggest claim 9 or claims dependent therefrom of the present application.

Reconsideration and withdrawal of this rejection are respectfully requested.

D. JP '821 in view of Sonobe et al.

Claims 1 and 7 were rejected under 35 U.S.C. §103(a) as allegedly being obvious over JP '821 in view of Sonobe et al. This rejection is respectfully traversed.

In light of the cancellation of claims 1 and 7, the rejection with respect to these claims is considered moot.

Reconsideration and withdrawal of this rejection are respectfully requested.

E. JP '821 in view of JP '410 and JP '159

Claim 3 was rejected under 35 U.S.C. §103(a) as allegedly being obvious over JP '821 in view of JP '410 and further in view of JP 63-218159 (JP '159). This rejection is respectfully traversed.

In light of the cancellation of claim 3, the rejection with respect to this claim is considered moot.

Reconsideration and withdrawal of this rejection are respectfully requested.

F. JP '821 in view of JP '410 and JP '378

Claim 5 was rejected under 35 U.S.C. §103(a) as allegedly being obvious over JP '821 in view of JP '410 and further in view of JP 2003-045378. This rejection is respectfully traversed.

In light of the cancellation of claim 5, the rejection with respect to this claim is considered moot.

Reconsideration and withdrawal of this rejection are respectfully requested.

G. JP '821 in view of JP '410 and JP '159

Claim 12 was rejected under 35 U.S.C. §103(a) as allegedly being obvious over JP '821 in view of JP '410 and further in view of JP '159. This rejection is respectfully traversed.

None of JP '821, JP '410 and JP '159, taken singly or in combination, teaches or suggests an electrochemical capacitor comprising a cathode that contains a fibrous carbon material as a constituent material, wherein the fibrous carbon material has a specific surface area of 2000 to 3000 m²/g as required by claim 12.

As set forth above with respect to claim 9, JP '821 does not teach that the fibrous carbon material has a specific surface area of 2000 to 3000 m²/g. Further, as explained above, JP '821 teaches away from the recited cathode of claim 12.

Even if JP '410 and JP '159 were to have been combined with JP '821, the subject matter of claim 12 would not have been achieved. JP '410 and JP '159 do not remedy the deficiencies of JP '821 detailed above. For example, like JP '821, nowhere do JP '410 and JP '159 disclose the cathode containing a fibrous carbon material that has the specific surface area of 2000 to 3000 m²/g as recited in claim 12.

For at least the foregoing reasons, Applicants respectfully submit that JP '821, JP '410 and JP '159, taken singly or in combination, fail to teach or suggest claim 12 of the present application.

Reconsideration and withdrawal of this rejection are respectfully requested.

II. New Claims

The references of record, taken singly or in combination, further fail to disclose an electrochemical capacitor comprising a cathode that contains a fibrous carbon material having an electronic conductivity as a constituent material, wherein the fibrous carbon material has a specific surface area of 2000 to 3000 m²/g as recited in amended claim 9, in addition to the further features of new claims 13-16.

III. Conclusion

In view of the foregoing, it is respectfully submitted that this application is in condition for allowance. Favorable reconsideration and prompt allowance of claims 9-16 are earnestly solicited.

Should the Examiner believe that anything further would be desirable in order to place this application in even better condition for allowance, the Examiner is invited to contact the undersigned at the telephone number set forth below.

Respectfully submitted,



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Attachment:

Translation of JP 60-211821

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(54) Title of the invention: Electric Double Layer Capacitor

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Specification

10 1. Title of the Invention

Electric Double Layer Capacitor

15 2. Claim(s)

(1) An electric double layer capacitor, having an electric double layer formed by a polarizable electrode body and an electrolyte interface, wherein the specific surface area of a positive electrode side of the polarizable electrode body is smaller than the specific surface area of a negative electrode side of the polarizable electrode body.

20 (2) The electric double layer capacitor according to Claim 1, wherein the pore size of the positive electrode side of the polarizable electrode body has a distribution where 10% or more are between 10 and 30 angstroms, and the pore size of the negative electrode side of the polarizable electrode body has a distribution where 70% or more are between 20 and 40 angstroms.

(3) The electric double layer capacitor according to Claim 1, wherein fibrous, papery, felt-like, or porous carbon or activated carbon is used as the polarizable electrode body.

5 (4) The electric double layer capacitor according to Claim 1, wherein an organic electrolytic solution is used as the electrolytic solution.

3. Detailed Description of the Invention

10 Industrial Field of Application

The present invention relates to a miniature high-capacitance wet type electric double layer capacitor.

15 Construction of a Conventional Example and Problems Thereof

Fig. 1 shows an example of the construction of the conventional electric double layer capacitor.

20 This example has a construction where an active carbon fiber sheet is used as a polarizable electrode body 1, and a metal layer such as aluminum or titanium or the like or a conductive resin layer is formed as a conductive electrode 2. These electrodes are overlaid with a separator 3 therebetween and after an electrolytic solution is added, are stored and sealed in a coin shaped case 5 in a state where the positive 25 and negative electrodes are insulated by a gasket 4. A metal conductive electrode 2 is formed by either a plasma spray

method or an arc spray method, or if a conductive resin is used, is formed by a conductive resin that primarily uses carbon as the conductive particles using either a screen printing method, spray method, or dip method.

5 If a conductive resin is used, the internal impedance will be higher than if a metal layer is used, and the capacitor will not be suitable for high discharge applications.

10 Conventionally, this type of capacitor uses (1) a water based electrolytic solution, and (2) a waterless electrolytic solution. The waterless system of (2), or in other words an organic electrolytic solution has lower conductivity than a water based electrolytic solution, but the dielectric strength is higher. The solvent can be propylene carbonate, gamma-butyl lactone, N-N-dimethylformamide, or acetonitrile 15 or the like. The solute can be a perchlorate of a tetraalkyl ammonium, such as tetraethyl ammonium perchlorate, or a hexafluorophosphate or fluoroborate of tetraalkyl ammonium, or a perchlorate of lithium, sodium, or potassium or the like. In particular, current practical use electric double layer 20 capacitors use activated carbon as the polarizable electrode.

If activated carbon is used as the polarizable electrode, the characteristics of the capacitor will be largely controlled by the following three items.

- (a) Specific surface area
- (b) Pore diameter
- (c) Pore volume

Furthermore the capacity of the electric double layer capacitor is expressed by equation (1).

$$\eta = d\phi/4\pi\delta \quad (1)$$

η : electric charge per unit area

d: dielectric constant of the medium

δ : average distance from the surface of the solid to the electrolyte ions

ϕ : double layer electrical potential

Therefore, if the amount of electrical charge that accumulates in a single cell is Q and the area formed by the double layer is S, Q can be expressed by Equation (2).

$$Q = dS\phi/4\pi\delta \quad (2)$$

Therefore, a larger surface area formed by the double layer in an electric double layer capacitor will result in a larger amount of accumulated electrical charge. Therefore, the specific surface area of condition (a) should be as large as possible. However, to increase the specific surface area, activation must be proceeded, and there are problems in that the mechanical strength will be greatly reduced.

Generally, when the specific surface area is increased, the pore volume (c) also increases. However, if the pore diameter (b) remains small as shown in Fig. 2, regardless of the extent of increase in the specific surface area, a double layer with good efficiency can not be formed. In the drawing, 6 represents a polarizable electrode, 6a represents a pore, and 6b represents the electrolyte ion. Based on the

principles of capillary condensation, in order to form a double layer with good efficiency, the pore diameter 8 must be 4 or more times the diameter 7 of the electrolyte ion as shown in Fig. 3. Particularly at low temperatures of 0°C or lower, the viscosity of organic solvents will increase, and water-based electrolytic solutions will begin to freeze, so the mobility of the electrolyte ion will be greatly diminished, and not only will formation of a double layer become difficult, but even a double layer which has previously been formed will not be easily discharged.

For the above reasons, a conventional electric double layer capacitor which uses activated carbon fiber as the polarizable electrode and an organic electrolytic solution as the electrolytic solution has an extremely large specific surface area of 2000 m²/g for both the positive and negative electrodes, and uses activated carbon fibers where a majority of the pore diameters are between 2 and 4 nm. However, activated carbon fibers which have these large characteristics have a disadvantage in that the activation efficiency is extremely low, at approximately 20%.

Object of the Invention

An object of the present invention is to provide an electric double layer capacitor with improved efficiency for forming an electric double layer per unit volume.

Construction of the Invention

In order to achieve this object, the present invention has a specific surface area of the positive electrode side of a polarizable electrode body that is smaller than the specific surface area of the negative electrode side of the polarizable electrode body.

Description of the Examples

Before describing specific examples, the activated carbon, carbon, and graphite used on the positive electrode side and the negative electrode side of the present invention and the electric double layer which is formed by the electrolyte ions will be described.

The effects of the present invention are more significant when an organic electrolytic solution is used rather than a water-based electrolytic solution, and when activated carbon fiber is used rather than activated carbon particles.

The reason for this is discussed below.

When an electrolyte such as tetraethyl ammonium perchlorate or lithium perchlorate or the like is dissolved in an organic solvent such as propylene carbonate or gamma-butyl lactone or the like, the perchlorate ions (ClO_4^-) have an ionic radius of 2.36 angstroms, so even though the lithium ions (Li^+) have a small ionic radius of 0.6 angstroms, generally the non-protonic polar solvent will more strongly

solvate cations than anions, so conversely, the ionic radius including the solvate of the cations will be larger than that of the anions. This condition is schematically showing in Fig. 4. 9 represents a solvated anion, 10 represents a solvated cation, 11 represents a positive electrode activated carbon fiber, 11' represents a negative electrode activated carbon fiber, and 6a represents a pore. The results shown in Table 1 can be achieved by proceeding with carbon activation using the same starting materials. When fabricating a coin shaped capacitor, these materials are stamped to have the same area, so when forming the positive electrode, or in other words the anion and the double layer, using stage (2) from Table 1 is best because the activation yield will be favorable, resistance will be low, and the strength will be high. However, for the negative electrode, sufficient activation must proceed as shown with stage (3), so activated carbon fiber with a large specific surface area and sufficiently large pore diameter is necessary so that the soluted cations can sufficiently penetrate into the pores.

Table 1

Stage	Progression of Activation	Specific Surface Area	Pore Volume	Pore Diameter	Strength
(1)	Progression	Small	Small	Small	Large
(2)		Remarkably Large	Large	Large	Small
(3)					

The state of the pores in activated carbon particles 12 and activated carbon fiber 11 are schematically shown in Fig. 5. As can be seen from the figure, activated carbon particles have micropores 14 inside macropores 13, so the penetration of the electrolyte is easier than with materials which have direct micropores 14 such as activated carbon fibers. Therefore, it is thought that if activated carbon particles are used for both the positive and negative electrodes, the effect of the present invention will not be significant.

As described above, the present invention is extremely effective as a system with a polarizable electrode body such as activated carbon fiber that can easily control the size of the pore diameter and an organic electrolytic solution which can strongly solvate cations. In other words, a material must be used where even cations which are strongly solvated can penetrate the pores of the negative electrode side of a polarizable electrode and form a double layer. However, for the positive electrode side of the polarizable electrode, a

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material is used where the carbon activation yield is higher than that of the negative electrode side so that penetration by the anions is possible, the strength is high, and the electrical resistance is low, and when combined with the negative electrode side of the polarizable electrode body, a double layer with the highest efficiency can be formed, raw materials can be efficiently used, and productivity can be increased.

Example 1

10 Phenol, acrylonitrile, and rayon fibers were carbonized and carbon activated, to obtain carbon fibers and activated carbon fibers which had the characteristics shown by (a) through (o) in Table 2. A collector body was formed from an aluminum layer of approximately 300 μm using a plasma spray method. Coin shaped capacitors as shown in Fig. 6
15 were manufactured using the various positive and negative electrode polarizable electrode bodies shown in Table 2 (a) through (o) in the combinations shown in Table 3, and the various characteristics are shown in the same table. In Fig. 6,
20 15 represents a positive electrode case, 16 represents a positive electrode side of a polarizable electrode with a collector body (aluminum) 16a, 17 represents a negative electrode case, 18 represents a negative electrode side polarizable electrode with a collector body (aluminum) 18a, 19 represents a separator, and 20 represents a gasket. The
25 electrolytic solution was an organic electrolytic solution where 1 mol of electrochemically stable tetraethyl ammonium

perchlorate was dissolved in a 1:1 solvent blend of propylene carbonate and gamma-butyl lactone. Each of the electrodes were stamped as a circle with a 14 mm diameter.

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Table 2

No.	Material	Specific Surface Area (BET Method) (m ² /g)	Pore Distribution	Mass per unit area (g/m ³)	Carbonized or carbon activation yield (%)
a	phenol fiber	2 to 10	Almost nonexistent	260	48
b	phenol fiber	500 to 600	10% or more between 10 and 30 angstroms	216	40
c	phenol fiber	900 to 1000	50% or more between 10 and 30 angstroms	200	37
d	phenol fiber	1500 to 1600	80% or more between 10 and 30 angstroms	180	33
e	phenol fiber	2000 to 2100	70% or more between 20 and 40 angstroms	113	21
f	phenol fiber	2400 to 2500	90% or more between 20 and 40 angstroms	92	17
g	polyacrylonitrile fiber	2 to 10	almost nonexistent	250	46
h	polyacrylonitrile	300 to	20% or more	140	26

	fiber	400	between 10 and 30 angstroms		
i	polyacrylonitrile fiber	700 to 800	70% or more between 20 and 40 angstroms	80	20
j	polyacrylonitrile fiber	1100 to 1200	90% or more between 20 and 40 angstroms	60	15
k	rayon fiber	2 to 10	almost nonexistent	255	47
l	rayon fiber	300 to 400	80% or more between 10 and 30 angstroms	85	16
m	rayon fiber	700 to 800	70% or more between 20 and 40 angstroms	65	12
n	phenol felt	900 to 1000	70% or more between 10 and 30 angstroms	100	--
o	polyacrylonitrile felt	300 to 400	60% or more between 10 and 30 angstroms	70	--

Table 3

No.	Electrode Construction		Properties		
	Positive electrode side polarizable electrode	Negative electrode side polarizable electrode	Capacitance (F)	Low temp. properties; -21°C capacitance/room temp. capacitance	Impedance (ohm) 1 KHz
1	a	e	0.2	0.9 to 1.0	3.1
2	b	e	0.8	0.9 to 1.0	3.3
3	c	e	1.1	0.9 to 1.0	3.5
4	d	e	1.2	0.9 to 1.0	4.2
5	c	c	1.4	0	4.0
6	d	d	1.6	0.01 to 0.1	4.3
7	e	e	1.1	0.9 to 1.0	4.5
8	b	f	0.7	0.9 to 1.0	4.6
9	c	f	0.9	0.9 to 1.0	4.7
10	d	f	0.8	0.9 to 1.0	4.6
11	k	m	0.7	0.9 to 1.0	4.5
12	i	l	0.8	0.9 to 1.0	4.5
13	i	j	0.7	0.9 to 1.0	4.2
14	i	i	0.7	0.9 to 1.0	4.3
15	l	l	0.6	0.9 to 1.0	4.3
16	m	m	0.6	0.9 to 1.0	4.4
17	n	m	0.5	0.9 to 1.0	4.6
18	o	l	0.3	0.9 to 1.0	4.7

Favorable capacitor characteristics were obtained for No. 1, and 2, 3, 4, 8, 9, 10, 11, 12, 13, 15, 17, and 18 in Table 3 by combining the carbon fibers and activated carbon fibers with the characteristics shown in Table 2 into electrodes in Table 3. In other words, a low impedance capacitor with sufficiently high capacity and favorable low temperature

characteristics can be obtained even without using negative and positive electrodes which were sufficiently activated as in No. 7. With No. 5 and 6, the solvated cations could not easily penetrate into the negative electrode side, and low temperature properties were extremely poor.

Example 2

The capacitor shown in Fig. 6 is configured by the employment as the negative electrode-side polarizable electrode of a material prepared by the addition of one part polyflon as a binder to 100 parts activated carbon particles of specific surface area $1400 \text{ m}^2/\text{g}$ of which 80% or more has a fine pore diameter of size 20 to 100A and pressing in a titanium net, and the employment of the activated carbon fibres of c and d of Table 2 as the positive electrode. Table 4 shows the characteristics of this capacitor. It is clear from Table 4 that the characteristics exhibited by the capacitor of the present embodiment are satisfactory.

Table 4

Electrode configuration		Characteristics		
Positive electrode	Negative electrode	Capacity (F)	Low-temperature characteristics -21°C capacity/Room temperature capacity	Impedance (Ω)
Table 1c	Granular	1.2	0.9 to 1.0	4.0

	activated carbon			
Table 1d	Granular activated carbon	1.1	0.9 to 1.0	4.2

Example 3

A large capacitor as shown in Fig. 7 and Fig. 8 was fabricated using the combination of No. 4 in Table 3. The size was 10 cm long and 5 cm wide. In the drawings, 21 represents a positive electrode lead, 22 represents a positive electrode, 23 represents a separator, 24 represents a negative electrode lead, 25 represents a negative electrode, 26 represents polyethylene laminate resin, and Fig. 8 is a cross section diagram for the case where Fig. 7 is cut along line a-a'. The characteristics of this capacitor are shown in Table 5. The electrolytic solution for this example was made by dissolving 1 mol of lithium perchlorate in a 1:1 solvent blend of propylene carbonate and gamma-butyl lactone.

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Table 5

Characteristics		
Capacitance (F)	Low temperature properties -21°C capacitance/room temp. capacitance	Impedance (ohms)
92	0.9 to 1.0	0.04

Effect of the Invention

As described above, the present invention can provide a

5 small size high capacitance electric double layer capacitor with low impedance by using an electrode that can form an electric double layer with good efficiency for both anions and cations for both the positive electrode and negative electrode of a polarizable electrode.

4. Brief Description of the Drawings

10 Fig. 1 is a partially cutaway front view diagram showing an example of a conventional electric double layer capacitor;

Fig. 2 through Fig. 5 are schematic diagrams showing the condition of electrolyte ions and pores in the polarizable electrode;

15 Fig. 6 is a cross section diagram showing an electrical double layer capacitor according to an example of the present invention;

Fig. 7 is a top view diagram showing an electric double layer capacitor according to another example of the present invention; and

20 Fig. 8 is a cross section diagram cut along line a-a' of Fig. 7.

25 16 POSITIVE ELECTRODE SIDE POLARIZABLE
ELECTRODE

18 NEGATIVE ELECTRODE SIDE

POLARIZABLE ELECTRODE

22 POSITIVE ELECTRODE

25 NEGATIVE ELECTRODE

5 Name of Agent Patent Attorney, Toshio Nakao and 1
other